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HOT WORKING OF HIGH-PURITY NICKEL-NIOBIUM ALLOYS (PREPRINT)

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14. ABSTRACT
The present work deals with the influence of niobium in solid solution on the dynamic recrystallization of pure nickel. High-purity nickel and two model nickel-niobium alloys were deformed to large strains via torsion at temperatures between 800 and 1000°C. Niobium additions considerably increased the flow stress, while they lowered the strain-rate sensitivity and increased the apparent activation energy. EBSD of the steady-state microstructures revealed strong grain refinement. Substructure development was favored, whereas thermal twinning was reduced by niobium. More generally, discontinuous recrystallization kinetics were considerably decreased.

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Nickel alloys, niobium, dynamic recrystallization, torsion testing

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Hot Working of High-Purity Nickel-Niobium Alloys

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Keywords: hot torsion, nickel, model materials, dynamic recrystallization, grain refinement

Abstract. The present work deals with the influence of niobium in solid solution on the dynamic recrystallization of pure nickel. High-purity nickel and two model nickel-niobium alloys were deformed to large strains via torsion at temperatures between 800 and 1000°C. Niobium additions considerably increased the flow stress, while they lowered the strain-rate sensitivity and increased the apparent activation energy. EBSD of the steady-state microstructures revealed strong grain refinement. Substructure development was favored, whereas thermal twinning was reduced by niobium. More generally, discontinuous recrystallization kinetics were considerably decreased.

Introduction

Dynamic recrystallization (DRX) has been much less investigated in nickel-base superalloys than in steels. This is mainly due to the fact that such alloys contain a large number of additional elements, which may act in a complex way during hot deformation. In particular, the behavior of solid-solution niobium atoms is questionable; in spite of their low bulk diffusion rate, Nb solutes are able to interact with grain boundaries and reduce considerably grain-boundary mobility. Model materials were thus used, including high purity nickel and two Ni-Nb grades, in the present work to investigate the influence of Nb solutes on DRX during large-strain torsion tests under strain rate and temperature conditions similar to those in hot forging. The rheological behavior and steady-state microstructures of the three alloys were compared and interpreted.

Preparation and Conversion of the Model Materials

High-purity nickel (Ni) and two Ni-Nb alloys, containing nominally 0.1 wt% (Ni-0.1Nb) and 1.0 wt% (Ni-1Nb) niobium, were prepared. For Ni, electrolytic-purity nickel (containing 150 ppm Fe, still present after purification) was melted by induction in a cold silver crucible under a high-purity argon-hydrogen atmosphere. For the two alloys, high-purity niobium was added to the melt [1]. The final Nb contents were 0.092 % and 0.953 %, respectively and the level of metalloids (C, S, O, N) was generally below 5 ppm. Each of the three ingots weighed ~1.2 kg and was converted by forging at 1050°C into bars of diameter 14 mm, which were then reduced to a 10-mm diameter by hot swaging. After homogenization heat treatment of 1 h at 700°C, the grain size of Ni and Ni-0.1 Nb was close to 500 µm, while it was much smaller (50-100 µm) in the Ni-1Nb grade. According to the Ni-Nb phase diagram, Nb is entirely in solid solution within the investigated hot working range.

Hot Torsion Stress-Strain Curves

Torsion was used to achieve large strains under well-controlled (radial) strain-gradient conditions up to the steady-state flow stress domain at temperatures ranging between 800 and 1000°C ($\approx$ 0.62-0.74 T_m) and three (von Mises equivalent) strain rates, *viz.*, 0.03, 0.10, and 0.30 s^{-1}. Specimens

were all quenched immediately after straining using a room-temperature flow of argon. Stress-strain curves are shown below for pure nickel (Fig. 1a) and the two Ni-Nb alloys (Fig. 1b, c) at the intermediate strain rate of 0.1 s^{-1}. There was no evidence of deformation heating at this strain rate.

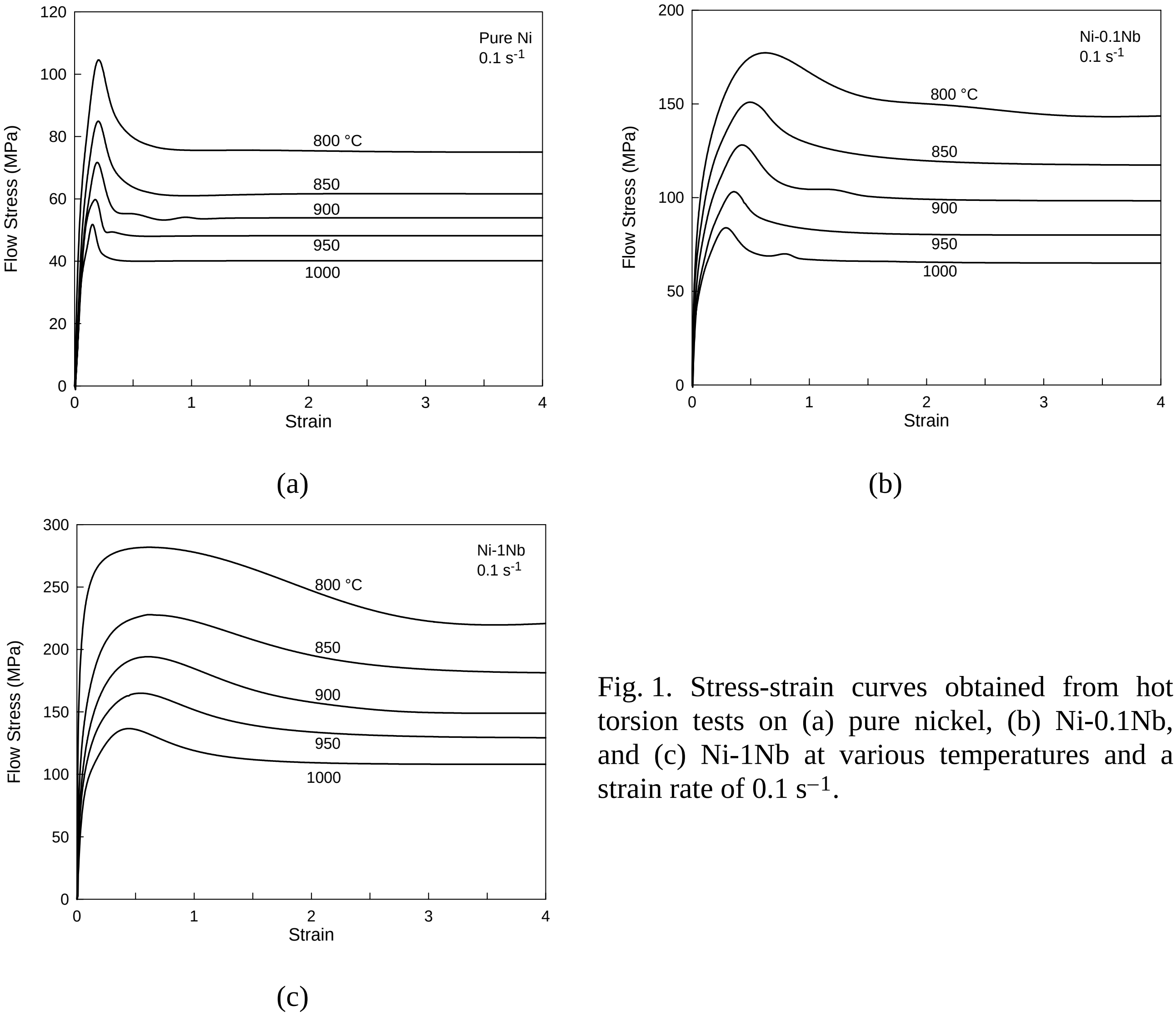

Fig. 1. Stress-strain curves obtained from hot torsion tests on (a) pure nickel, (b) Ni-0.1Nb, and (c) Ni-1Nb at various temperatures and a strain rate of 0.1 s^{-1}.

The data revealed that the overall stress level was noticeably increased by Nb in solid solution. This was described empirically by a simple power-law equation:

$$\sigma = \sigma_0 + (\sigma_1 - \sigma_0)\, x^p, \tag{1}$$

in which x is the Nb content (wt%), and σ_0 and σ_1 denote the flow stresses of pure nickel and Ni-1Nb, respectively. Eq. 1 holds for both the peak and steady-state stresses; the values of exponent p were quite similar whatever the temperature, with an average of 0.38.

The shape of the flow curves was also strongly affected by the addition of niobium. For pure nickel, the steady-state flow stress was attained at strains of ~0.5 at 1000°C to 1.0 at 800°C, strains of 1 to 3 for Ni-0.1Nb, and strains of 2 to 3.5 for Ni-1Nb. All the flow curves exhibited single-peak behavior classically associated with discontinuous dynamic recrystallization; the slight irregular oscillations observed for some of the curves for Ni and Ni-0.1Nb were ascribed to experimental artifacts linked for instance to the large initial grain sizes. The present observations for high-purity nickel are in agreement with the results of Luton and Sellars [2]. These authors observed multiple-peak flow curves only when the large strain flow stress level was less than about 41 MPa, which

corresponds to the minimum steady-state flow stress reached in the present investigation (at 1000°C). For the Ni-Nb alloys, it is not surprising that multiple-peak flow curves were not observed, because niobium in solid solution is expected to decrease grain-boundary mobility, and thus to limit the increased grain size normally associated with wavy flow curves.

Strain Rate and Temperature Dependence of the Flow Stress

The strain rate sensitivity parameter $m = \partial \ln \sigma / \partial \ln \dot{\varepsilon}|_{T,\varepsilon}$ for the three materials was observed to increase slightly with temperature between 800 and 1000°C. The dependence of m on niobium content at 900°C is displayed in Fig. 2 for the maximum stress (which may not occur at constant strain strictly speaking) and the steady-state flow stress. The m values were much smaller for Ni-0.1Nb than for Ni, but further addition of niobium up to 1 % led to a limited decrease of m.

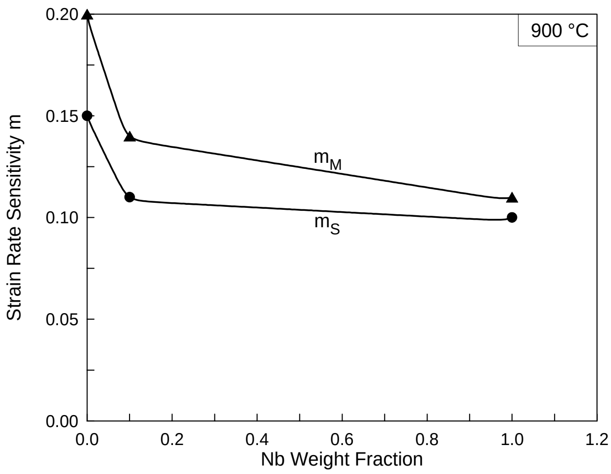

Fig. 2. Influence of niobium content on the strain-rate sensitivities for the maximum (m_M) and steady-state (m_S) flow stresses at 900°C.

The apparent activation energies of hot deformation were measured at $\dot{\varepsilon} = 0.1\,s^{-1}$ for both the peak and steady-state stresses from $\ln \sigma - 1/T$ diagrams, where T is the absolute deformation temperature. The data were perfectly fit by straight lines in all cases. Thus, following the classical analysis of the strain-rate and temperature dependence of flow stress, this means that $mQ/R = \partial \ln \sigma / \partial (1/T)|_{\dot{\varepsilon},\varepsilon}$ was independent of temperature, where R is the gas constant. The resulting activation energies Q_M and Q_S, pertaining to the maximum and steady state flow stresses, respectively, are plotted in Fig. 3 as a function of the niobium content. For pure Ni, $Q_M \approx 200$ kJ/mol is significantly less than the value of 234 kJ/mol found by Luton and Sellars [2]. For the two Ni-Nb alloys, Q_M and Q_S were much larger than the activation energies of pure nickel, but the main increase was produced by the presence of 0.1 %Nb, whereas the further addition of niobium to 1 % had much less effect on the level of Q.

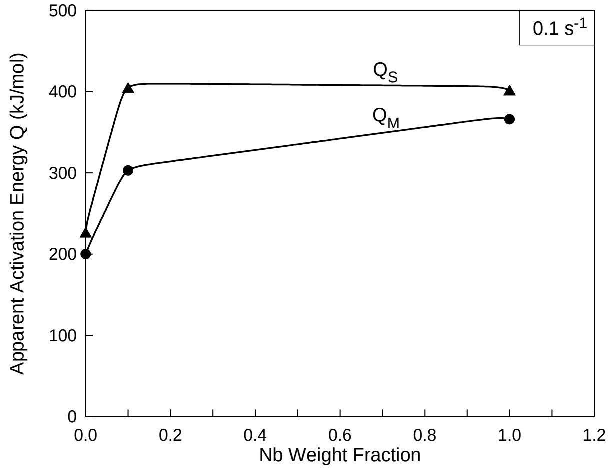

Fig. 3. Influence of niobium content on the apparent activation energies Q_M and Q_S at 0.1 s^{-1} determined from the peak stress and steady-state stress, respectively.

Hot Deformation Microstructures

Microstructure and local texture investigations were carried out by electron backscatter diffraction (EBSD) on torsion specimens of each of the three materials deformed under the same conditions, *viz.*, 900°C, 0.1 s^{-1} and a strain of 5, associated with steady-state flow stress (but not necessarily steady-state microstructure and texture). In Figures 4a-c, special boundaries are indicated by different colors: low-angle boundaries (LAB) are identified by a range of gray → pink → red lines corresponding to misorientation angles increasing from 2 deg (detection limit) to 15 deg. Ordinary high-angle boundaries (HAB) are shown in black. Green lines correspond in turn to twin boundaries (TB), *i.e.*, boundaries associated with a rotation of 60 deg around a <111> crystallographic axis. A more quantitative description of the fractional areas associated with the various types of boundary is given by the misorientation-angle distributions (Figure 4d-f). The various microstructural parameters are summarized in Table 1.

The results in Fig. 4 and Table 1 revealed that the addition of niobium favors substructure development, as shown by the fractions of LABs which increased from ~0.02 for pure nickel, to 0.16 for Ni-0.1Nb, and 0.32 for Ni-1Nb. This may be due to the stabilization of dislocation walls by niobium solutes (*i.e.*, decrease of dynamic-recovery kinetics) and/or the decrease of grain-boundary mobility, because substructure is usually swept out by moving grain boundaries.

By contrast, the twin-boundary area fraction (here, by convention, boundaries with $59 < \theta < 61$ deg) was strongly reduced by niobium, decreasing from 0.50 for pure nickel, to 0.28 for Ni-0.1Nb and 0.07 for Ni-1Nb at 900°C. The value of 0.50 for Ni is characteristic of a statically fully-recrystallized, low-stacking-fault-energy metal [3], which suggests that the microstructure in Fig. 4a had undergone some post-dynamic evolution before quenching. The reduction of twinning by solutes has been observed previously in other systems such as Cu-Sn alloys [4]. It may be attributed to the decrease of grain-boundary mobility because thermal twinning during deformation is associated with grain-boundary migration [5].

Finally, the grains appeared to be randomly oriented in deformed pure nickel, as a result of discontinuous DRX. By contrast, a strong texture was apparent for Ni-0.1Nb, and became very pronounced for Ni-1Nb. The main components belonged to the {111}<uvw> fiber. (Here, {hkl} denotes the shear plane perpendicular to the z-axis, and <uvw> the shear direction parallel to the θ axis). Depending on the area of investigation and the temperature, they seem to oscillate between two orientations: A_1^*/A_2^* $\{111\}<11\overline{2}>$, where each of the two orientations A_1^* and A_2^* is *self-symmetric* in the sense that it fulfills by itself the symmetry requirements of the torsion test, and

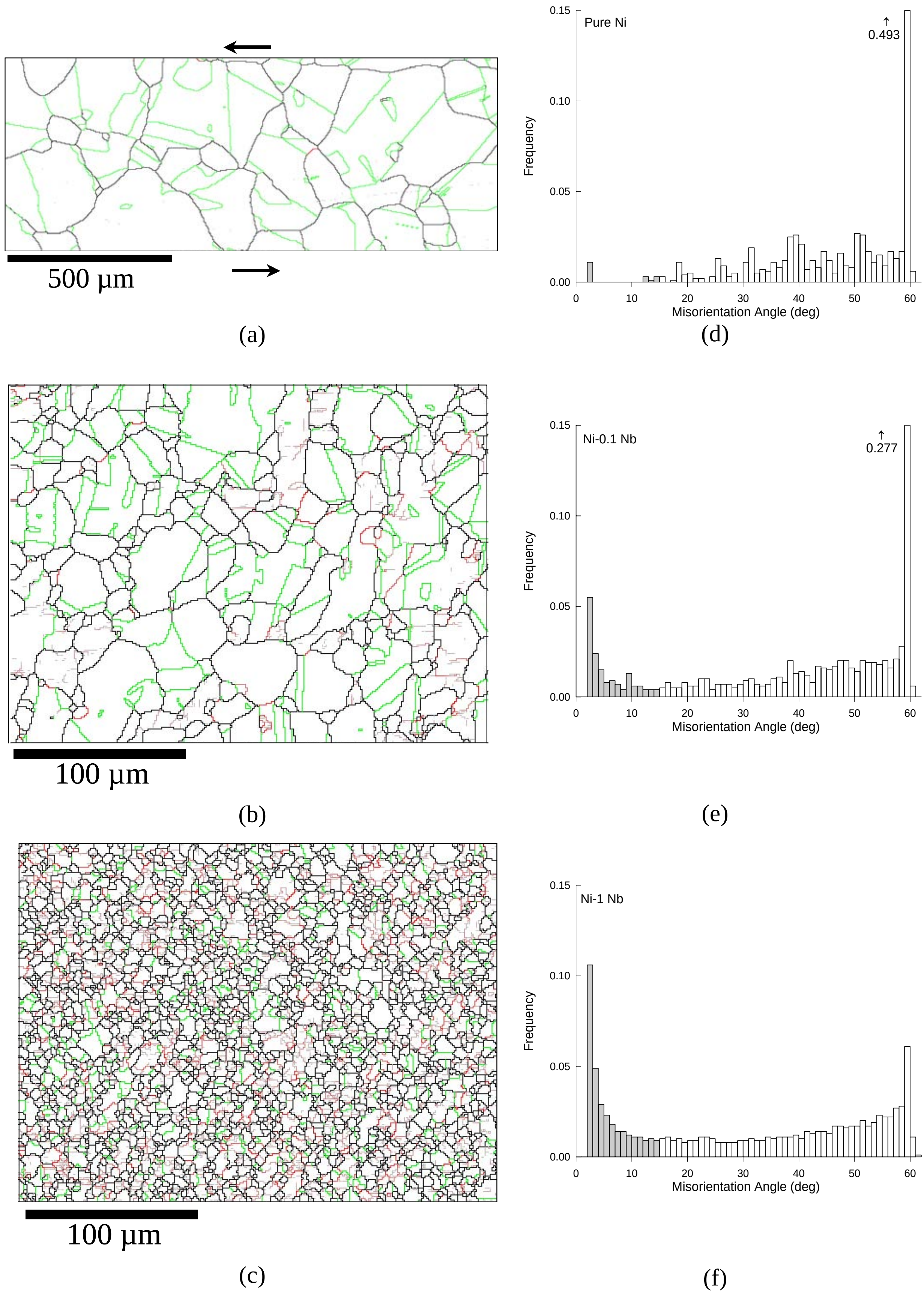

Fig. 4. (a, b, c) EBSD maps and (d, e, f) misorientation angle distributions in (a, d) pure Ni, (b, e) Ni-0.1Nb, and (c, f) Ni-1Nb after torsion at 900°C and 0.1 s^{-1} to a strain of 5. Arrows indicate the shear direction. Black: ordinary HABs; green: twin boundaries; grey-pink-red: LABs. Subgrain boundaries are associated with gray bars in the histograms.

Table 1. Influence of Nb content and deformation temperature on the quantitative microstructure parameters

	Ni 900°C	Ni-0.1Nb 900°C	Ni-1Nb 800°C	Ni-1Nb 900°C	Ni-1Nb 1000°C
HAB intercept [µm]	152	16	4	6	13
LAB fraction	0.02	0.16	0.33	0.32	0.32
TB fraction	0.50	0.28	0.05	0.07	0.13

$A/\bar{A}\,\{111\} < 1\bar{1}0 >$, in which A and $\bar{A}$ must coexist for symmetry (*twin-symmetric* orientation). It should be noted, however, that EBSD texture measurement on limited areas might not reflect the average textures of the specimens. It is nevertheless remarkable that the above orientations are invariant by twinning on the {111} plane that coincides with the shear plane, which may explain their resistance to DRX.

Conclusions

1) Stress-strain curves suggest that hot deformation of pure nickel, Ni-0.1 %Nb, and Ni-1 %Nb is associated with discontinuous dynamic recrystallization. With respect to pure nickel, the kinetics of flow softening are decreased by niobium solutes.
2) The product mQ of the strain-rate sensitivity and the apparent activation energy of deformation is remarkably independent of temperature, while m slightly increases (and therefore Q decreases) with test temperature within the investigated range 800-1000°C.
3) The strain-rate sensitivity m is significantly reduced by the presence of niobium in solid solution, whereas Q is increased. These variations with Nb content are more rapid between 0 and 0.1Nb than with further Nb additions, thus suggesting that a saturation effect occurs.
4) Deformation microstructures associated with the steady-state flow stress ($\varepsilon = 5$) are considerably refined by niobium solutes. Furthermore, the addition of Nb favors substructure development while it reduces thermal twinning during deformation.
5) Whereas deformation texture is eliminated by DRX in pure nickel, well developed components of the {111}<uvw> family form in the Nb-containing alloys. They are partially stable with respect to twinning and are therefore able to resist DRX.

Further investigations involving model Ni-Nb alloys with lower or higher niobium additions (and thus Ni_3Nb precipitates) will now be carried out with the aim of better understanding and controlling the hot working of Ni-base superalloys.

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